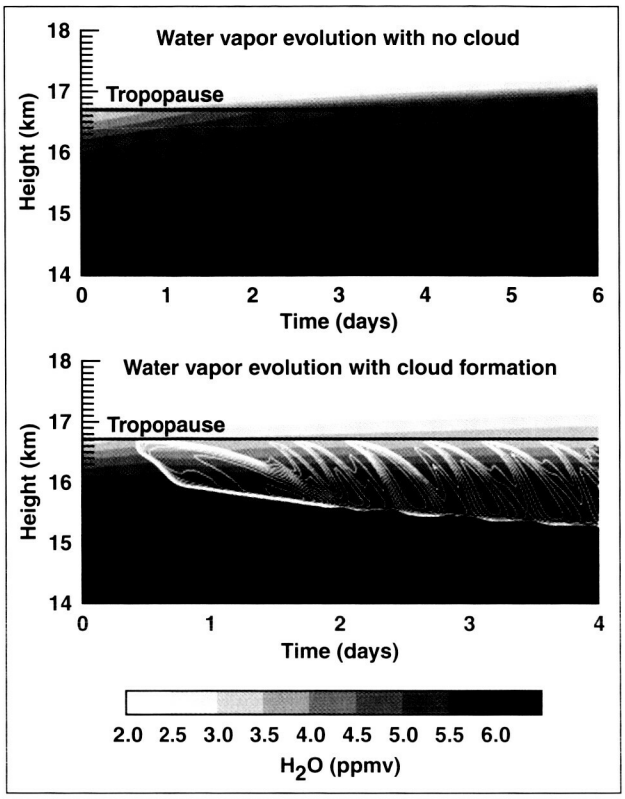

Fig. 1. Contours of water vapor mixing ratio (shading) are plotted versus time and height. The top panel shows that if no cloud forms, ascending air transports excessive amounts of water across the tropopause into the stratosphere. The bottom panel shows the dehydration of rising air if cloud formation is included in the model. The white contours show the cloud ice water content.

examples of supersaturated air near the tropopause (as predicted by the model); however, further observations of water vapor and wave motions near the tropical tropopause are required to clarify the cloud formation and dehydration process.

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Lofting of Soot Particles into the Middle Atmosphere by Gravito-Photophoresis

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The observed existence of soot aerosol at 20-kilometer (km) altitude (which arguably is generated by aircraft flying in corridors at 10–12 km) requires a transport mechanism in a thermally stable stratosphere that is different from isentropic or dynamic mixing. Such a mechanism could be provided by gravito-photophoresis, induced by the incidence of sunlight on strongly absorbing fractal soot particles. This particle absorptivity, in conjunction with uneven surface-coating of sulfuric acid and their fractal nature, makes soot particles (with maximum dimensions approaching 1 micrometer) particularly conducive to gravito-photophoresis. It is the requirement of a restoring torque that orients the particle with respect to gravity. This required force is provided by the fractal characteristics of soot, and a body-fixed photophoretic force is given by asymmetric thermal accommodation coefficients across the uneven surface of the particle.

During the Subsonic Assessment (SASS) Ozone and Nitrogen Oxides Experiment (SONEX) field campaign in 1997, soot aerosol was sampled in commercial airline flight corridors over the northeastern Atlantic, and the gravitational and gravito-photophoretic forces acting on those soot particles were computed. The result is that 16% by number, corresponding to 51% by mass, of a soot particle size distribution could be lofted against gravity by gravito-photophoresis. The calculated vertical velocities, exceeding settling velocities by up to a factor of 30, suggest that it takes about 30 years to transport soot from 10 to 20 km and 20 years to transport soot from 20 to 80 km. On the basis of current stratospheric soot loading, the resulting soot mass flux at 20-km altitude is 5e-18 grams per square centimeter per second (g cm^{-2} sec^{-1}), which is within one order of magnitude of the influx of meteoritic dust into the mesosphere from outer space.

The effect of gravito-photophoresis is strongly altitude dependent. With increasing pressure near the Earth's surface, the lofting force falls off quickly. Above the mesopause, the lofting force becomes smaller because of a dominating energy loss by

radiation rather than by molecular heat transfer. Thus, gravito-photophoretic lofting forces are most effective within the altitude range between 10 and 85 km, making aircraft soot emitted in commercial flight corridors subject to lofting up to the mesosphere.

The current mass loading of soot in the middle atmosphere is too small to cause a direct absorption effect that would be comparable to the extinction of light by scattering on mesospheric cloud particles. However, it is conceivable that soot in the mesosphere has indirect effects by providing freezing nuclei for mesospheric ice particles to form. In addition, soot might contribute to the ionization of the mesosphere to affect the appearance of polar mesopheric summer radar echoes.

Collaborators in this research include H. Rohatschek (University of Linz, Austria), Sunita Verma (Science Systems & Application, Inc.), Nina Boiadjieva (San Jose State University), and Steve Howard (Symtech Corporation).

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Physical and Chemical Properties of Aerosols and Cloud Particles

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Recent modeling studies have suggested a link between black carbon aerosol (BCA) and ozone chemistry via the reduction of nitric acid, nitrogen dioxide, and ozone on BCA particles. The ozone reaction converts ozone to oxygen molecules, while nitric acid can react to form nitrogen oxide (NO_x). Also, a buildup of BCA could reduce the single-scatter albedo of aerosol below a value of 0.98, a critical value that has been postulated to change the radiative effect of stratospheric aerosol from cooling to warming. Correlations between measured BCA amounts and aircraft usage have been reported. Attempts to link BCA to ozone chemistry and other stratospheric processes have been hindered by questions concerning the amount of BCA that exists in the stratosphere, the magnitude of reaction probabilities, and the scarcity of BCA measurements. The primary objectives of the Ames team (as part of the Photochemistry of Ozone Loss in the Arctic Region in Summer (POLARIS) mission) were (1) to determine the distribution of sulfate and soot aerosols, and (2) to determine the role of these aerosols in stratospheric photochemistry.

To facilitate measurements, a system that automates the operation of the Ames Wire Impactors (AWI) was designed, fabricated, and flown successfully on POLARIS. This system alleviates the pilot workload and allows the experimenter the flexibility to sample at predetermined altitudes, locations, or temperatures.

Because of the fractal nature of BCA, modification of the AWI data-analysis procedures was required in which the collection of BCA is modeled as a fractal aggregate. The new method results in an increase in the measured BCA surface area of about 15 times and an increase in soot loading of about 6 times over the previously used approach. Despite this increase, BCA surface area is only about 10% of the measured sulfuric acid aerosol surface area.

Including heterogeneous reactions on BCA in a photochemical model can affect photochemistry, leading to renoxification and increased ozone depletion. However, these predicted effects are not supported by the POLARIS observations, in particular the nitrogen oxide/reactive nitrogen (NO_x/NO_y) ratios. Including BCA reactions does not statistically improve the agreement between model and measurement in any of several scenarios considered. Furthermore, if the reactions cause even partial carbon oxidation, the BCA would be consumed at a rate inconsistent with POLARIS observations. These inconsistencies lead to the conclusion that the presence of BCA in the stratosphere did not affect stratospheric photochemistry during POLARIS.

Measurements of aircraft plumes indicate that aircraft emit substantial numbers of volatile and nonvolatile particles during flight. These observations have caused concern that commercial aviation sources may significantly influence heterogeneous processes, cloud formation, and microphysics in the